

D-wave-like nodal superconductivity in the organic conductor (TMTSF)₂ClO₄

A.G. Lebed *

*Department of Physics, University of Arizona, 1118 E. 4th Street, Tucson, Arizona 85721, USA
L.D. Landau Institute for Theoretical Physics, 2 Kosygina Street, 117334 Moscow, Russia*

Abstract

We suggest theoretical explanation of the high upper critical magnetic field, perpendicular to conducting chains, $H_{c2}^{b'}$, experimentally observed in the superconductor (TMTSF)₂ClO₄, in terms of singlet superconducting pairing. In particular, we compare the results of d-wave-like nodal, d-wave-like node-less, and s-wave scenarios of superconductivity. We show that, in d-wave-like nodal scenario, superconductivity can naturally exceed both the orbital upper critical magnetic field and Clogston-Shandrasekhar paramagnetic limit as well as reach experimental value, $H_{c2}^{b'} \simeq 6 T$, in contrast to d-wave-like node-less and s-wave scenarios. In our opinion, the obtained results are strongly in favor of d-wave-like nodal superconductivity in (TMTSF)₂ClO₄, whereas, in a sister compound, (TMTSF)₂PF₆, we expect either the existence of triplet order parameter or the coexistence of triplet and singlet order parameters.

Key words: organic superconductor, upper critical magnetic field
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1. Introduction

High magnetic field properties of (TMTSF)₂X (X=ClO₄, PF₆, etc.) organic materials have been intensively studied [1] since the discovery of superconductivity in (TMTSF)₂PF₆ conductor [2]. From the beginning, it was clear that superconductivity in the above mentioned materials was unconventional. Indeed, early experiments [3,4] showed that the Hebel-Slichter peak was absent in the NMR measurements [3] and that superconductivity is destroyed by non-magnetic impurities [4]. These facts were strong arguments that superconducting order parameter changed its sign on the quasi-one-dimensional (Q1D) Fermi surfaces (FS) of (TMTSF)₂X material. The main results of both experiments [3,4] were recently confirmed in a number of publications (see, for example, [5,6]). It is important that the above mentioned experiments

did not contain information about spin part of superconducting order parameter and could not distinguish between singlet and triplet superconducting pairings.

The first measurements of the Knight shift in (TMTSF)₂PF₆ conductor [6,7] showed that it was not changed in superconducting phase, which was interpreted in favor of triplet superconductivity [6,7]. On the other hand, the more recent Knight shift measurements [8] in superconductor (TMTSF)₂ClO₄ have shown a clear change of the Knight shift in superconducting phase at relatively low magnetic fields, $H \simeq 1 T$, and have been interpreted as evidence of singlet superconductivity [8]. Another argument in favor of singlet order parameter is the fact that the upper critical magnetic field, parallel to conducting axis, H_{c2}^a [9], is paramagnetically limited [10]. Moreover, very recently the Larkin-Ovchinnikov-Fulde-Ferrell phase [11,12], which appears for singlet superconducting pairing, has been experimentally discovered [13,14] in (TMTSF)₂ClO₄ and theoretically interpreted [15].

* Tel: +1 520 626 1031; FAX: +1 520 621 4721; e-mail: lebed@physics.arizona.edu

2. Goal

In such situation, where support of singlet superconducting pairing in (TMTSF)₂ClO₄ material is increasing, it is important to reinvestigate theoretically high experimental upper critical magnetic fields, $H_{c2}^{b'}$ [13,14,16], observed for a magnetic field, perpendicular to conducting chains. For many years, large values of $H_{c2}^{b'}$ have been considered as a consequence of triplet superconducting pairing. Our goal is to show that we can naturally explain large values of $H_{c2}^{b'}$ within singlet d-wave-like nodal scenario of superconductivity in (TMTSF)₂ClO₄. We also show that d-wave nod-less and s-wave scenarios are much less consistent with experimental value of $H_{c2}^{b'} \simeq 6$ T [13,14]. This value exceeds both the quasi-classical upper critical field [17] and Clogston-Shandrasekhar paramagnetic limit [18] due to the coexistence of two unusual superconducting phases: Reentrant superconductivity [19-24] and Larkin-Ovchinnikov-Fulde-Ferrell phase [11,12].

3. Results

Let us consider Q1D spectrum of (TMTSF)₂ClO₄ conductor in tight binding model [1],

$$\epsilon(\mathbf{p}) = -2t_a \cos(p_x a/2) - 2t_b \cos(p_y b') - 2t_c \cos(p_z c^*), \quad (1)$$

in a magnetic field, perpendicular to its conducting chains,

$$\mathbf{H} = (0, H, 0), \quad \mathbf{A} = (0, 0, -Hx), \quad (2)$$

where $t_a \gg t_b \gg t_c$ are electron hopping integrals along \mathbf{a} , \mathbf{b}' , and \mathbf{c}^* axes, respectively. Electron spectrum (1) can be linearized near two slightly corrugated sheets of Q1D FS as

$$\delta\epsilon^\pm(\mathbf{p}) = \pm v_x(p_y)[p_x \mp p_F(p_y)] - 2t_c \cos(p_z c^*), \quad (3)$$

where $+$ ($-$) stands for right (left) sheet of Q1D FS.

We represent electron wave functions in a real space in the following way:

$$\begin{aligned} \Psi_\epsilon^\pm(x, y, z, \sigma) = & \exp[ip_F(p_y)x] \exp(ip_y y) \exp(ip_z z) \\ & \times \psi_\epsilon^\pm(x, p_y, p_z, \sigma). \end{aligned} \quad (4)$$

Let us use the Peierls substitution method,

$$p_x \mp p_F(p_y) \rightarrow -id/dx, \quad p_z \rightarrow p_z - eA_z/c. \quad (5)$$

As a result, the Schrodinger-like equation for wave functions $\psi^\pm(x, p_y, p_z, \sigma)$ can be written as

$$\begin{aligned} \left[\mp iv_x(p_y) \frac{d}{dx} - 2t_c \cos\left(p_z c^* + \frac{\omega_c}{v_F} x\right) - \mu_B \sigma H \right] \\ \times \psi_\epsilon^\pm(x, p_y, p_z, \sigma) = \delta\epsilon \psi_\epsilon^\pm(p_x, p_y, p_z, \sigma) \end{aligned} \quad (6)$$

where μ_B is the Bohr magneton, $\sigma = \pm 1$ stands for spin up and down, respectively; $\omega_c = eHv_F c^*/c$, $\delta\epsilon = \epsilon - \epsilon_F$.

It is important that Eq.(6) can be analytically solved:

$$\begin{aligned} \psi_\epsilon^\pm(x, p_y, p_z, \sigma) = & \frac{\exp[\pm i\delta\epsilon x/v_x(p_y)]}{\sqrt{2\pi v_x(p_y)}} \exp\left[\pm i \frac{\mu_B \sigma H x}{v_x(p_y)}\right] \\ & \times \exp\left[\pm i \frac{2t_c}{v_x(p_y)} \int_0^x \cos\left(p_z c^* + \frac{\omega_c}{v_F} u\right) du\right]. \end{aligned} \quad (7)$$

The corresponding Green functions can be obtained from the following equation (see Ref.[25]):

$$G(x, x_1, p_y, p_z, \sigma) = \sum_\epsilon \frac{\psi_\epsilon^*(x, p_y, p_z, \sigma) \psi_\epsilon(x_1, p_y, p_z, \sigma)}{i\omega_n - \epsilon}. \quad (8)$$

Below, we introduce superconducting order parameter in the following way:

$$\Delta(p_y, x) = f(p_y b') \Delta(x), \quad \int_0^{2\pi} f^2(p_y b') d(p_y b')/2\pi = 1, \quad (9)$$

where function $f(p_y b')$ takes into account three possible order parameters: d-wave-like nodal, $f(p_y b') = \sqrt{2} \cos(p_y b')$, s-wave, $f(p_y b') = 1$, and d-wave-like node-less, $f(p_y b') = 2\theta(p_y b' + \pi/2) - 2\theta(p_y b' - \pi/2) - 1$ [26]. Linearized gap equation for all three possible singlet scenarios of superconductivity in (TMTSF)₂ClO₄ conductor can be derived, using the Gor'kov equations [25] for non-uniform superconductivity [28-30]. As a result of rather lengthy but straightforward calculations, we obtain:

$$\begin{aligned} \Delta(x) = & \tilde{g} \int \frac{dp_y}{v_x(p_y)} \int_{|x-x_1| > \frac{v_x(p_y)}{\Omega}} \frac{2\pi T dx_1}{v_x(p_y) \sinh\left[\frac{2\pi T |x-x_1|}{v_x(p_y)}\right]} \\ & \times J_0 \left\{ \frac{8t_c v_F}{\omega_c v_x(p_y)} \sin\left[\frac{\omega_c(x-x_1)}{2v_F}\right] \sin\left[\frac{\omega_c(x+x_1)}{2v_F}\right] \right\} \\ & \times \cos\left[\frac{2\beta \mu_B H(x-x_1)}{v_x(p_y)}\right] f^2(p_y b') \Delta(x_1), \end{aligned} \quad (10)$$

where \tilde{g} is effective electron coupling constant, Ω is cut-off energy, parameter β takes into account possible deviations of superconductivity in (TMTSF)₂ClO₄ from weak coupling scenario.

Note that Eq.(10), derived in this article, is very general. It takes into account both the orbital and paramagnetic effects against superconductivity. In particular, it takes into account quantum nature of electron motion along open orbits in the extended Brillouin zone in Q1D conductor (3) in a magnetic field (2). It is possible to make sure that the main quantum parameter in Eq.(10) is $2t_c v_F / \omega_c v_x(p_y) \simeq 2t_c / \omega_c$. Let us

estimate the value of this quantum parameter, using quasi-classical language. In accordance with Ref.[19], quasi-classical electron trajectory in a magnetic field (2) can be written as

$$z(t, H) = c^* l_{\perp}(H) \cos(\omega_c t), \quad (11)$$

where $l_{\perp}(H) = 2t_c/\omega_c$ corresponds to a "size" of electron trajectory in terms of interlayer distance, c^* ; t is time.

It is possible to show that

$$l_{\perp}(H) = \frac{2\sqrt{2}}{\pi} \frac{\phi_0}{ac^*H} \frac{t_c}{t_a} \simeq \frac{2 \times 10^3}{H(T)} \frac{t_c}{t_b} \frac{t_b}{t_a}, \quad (12)$$

where $H(T)$ is a magnetic field, measured in Teslas, ϕ_0 is the flux quantum. Note that value of $t_a/t_b \simeq 10$ is very well known in (TMTSF)₂ClO₄ from theoretical fitting [31] of the so-called Lee-Naughton-Lebed oscillations [31,32]. As to ratio t_c/t_b , it can be evaluated from the measured Ginzburg-Landau (GL) slopes of the upper critical magnetic fields in (TMTSF)₂ClO₄ conductor [13,14]:

$$t_c/t_b = (b^*/\sqrt{2}c^*)(H_{c2}^c/H_{c2}^{b'})_{GL}, \quad (13)$$

$$t_c/t_b = (b^*/c^*)(H_{c2}^c/H_{c2}^{b'})_{GL}, \quad (14)$$

where Eq.(13) is valid for d-wave-like nodal pairing, whereas Eq.(14) is valid for both d-wave-like node-less and s-wave pairings.

As a result, we obtain

$$l_{\perp}(H = 6 T) \simeq 0.48 \quad (15)$$

for d-wave-like nodal scenario of superconductivity and

$$l_{\perp}(H = 6 T) \simeq 0.68 \quad (16)$$

for d-wave-like node-less and s-wave ones. Low values of the parameter $l_{\perp}(H = 6 T)$ show that both cases correspond to $3D \rightarrow 2D$ dimensional crossover of electron motion [19], where electrons are almost localized on conducting layers. In this situation, superconductivity becomes almost two-dimensional and we can approximate the Bessel function in Eq.(10) as $J_0(z) \simeq z^2/4$.

Below we consider the gap equation (10) at zero temperature, $T = 0$. In this case and under condition of $3D \rightarrow 2D$ dimensional crossover, it is possible to represent superconducting order parameter in the following way:

$$\Delta(x) = \exp(ikx)[1 + \alpha_1 \cos(2\omega_c x/v_F) + \alpha_2 \sin(2\omega_c x/v_F)], \quad (17)$$

where $|\alpha_1|, |\alpha_2| \ll 1$. For such order parameter Eq.(10) can be rewritten as

$$\frac{1}{g} = \int_0^{2\pi/b'} \frac{d(p_y b')}{2\pi} \int_{\frac{v_F}{\Omega}}^{\infty} \frac{dz}{z} f^2(p_y b') \cos\left(\frac{2\beta\mu_B H z}{v_F}\right) \times \frac{v_F}{v_x(p_y)} \left[1 - 2l_{\perp}^2(H) \sin^2\left(\frac{\omega_c z}{2v_F}\right)\right] \cos\left[\frac{v_x(p_y)}{v_F} k z\right] \quad (18)$$

where g is renormalized electron coupling constant, $x_1 - x = z v_x(p_y)/v_F$. Here, we consider Eq.(18), taking into account that electron velocity component along conducting \mathbf{x} axis is

$$v_x(p_y) = v_F[1 + \alpha \cos(p_y b')], \quad (19)$$

where $\alpha = \sqrt{2}t_b/t_a \simeq 0.14$ [23]. Under condition $\alpha \ll 1$, Eq.(18) can be simplified:

$$\frac{1}{g} = \int_{\frac{v_F}{\Omega}}^{\infty} \frac{dz}{z} \cos\left(\frac{2\beta\mu_B H z}{v_F}\right) \cos(kz)[J_0(\alpha kz) - m J_2(\alpha kz)] \times \left[1 - 2l_{\perp}^2(H) \sin^2\left(\frac{\omega_c z}{2v_F}\right)\right], \quad (20)$$

where $m = 1$ for d-wave-like nodal superconducting order parameter, whereas $m = 0$ for d-wave-like node-less and s-wave ones. It is important that, in the absence of the paramagnetic effects, Eq.(20) describes the Reentrant superconductivity [19] with transition temperature being increasing function of a magnetic field. Therefore, we call superconducting phase, described by Eq.(20), the hidden Reentrant superconductivity.

We can simplify Eq.(20) by using the following relationship:

$$\frac{1}{g} = \int_{\frac{v_F}{\Omega}}^{\infty} \frac{2\pi T_c dz}{v_F \sinh\left(\frac{2\pi T_c z}{v_F}\right)}, \quad (21)$$

where T_c is superconducting transition temperature in the absence of a magnetic field. As a result, we obtain

$$\ln\left(\frac{H_{c2}^{b'}}{H^*}\right) = \int_0^{\infty} \frac{dz}{z} \cos\left(\frac{2\beta\mu_B H z}{v_F}\right) \times \left\{ \cos(kz)[J_0(\alpha kz) - m J_2(\alpha kz)] \times \left[1 - 2l_{\perp}^2(H) \sin^2\left(\frac{\omega_c z}{2v_F}\right)\right] - 1 \right\}, \quad (22)$$

where $\mu_B H^* = \pi T_c/2\gamma$, γ is the Euler constant. We numerically find maxima of $H_{c2}^{b'}(k)$ as a function of wave vector, k , of the order parameter (17) for $m=1$ and $m=0$. We come to the conclusion that experimental value, $H_{c2}^{b'} \simeq 6 T$, can be obtained for d-wave-like

nodal order parameter at $\beta \simeq 0.85$, whereas for d-wave-like node-less and s-wave order parameters it corresponds $\beta \simeq 0.5$. Note that the so-called g -factor in $(\text{TMTSF})_2\text{ClO}_4$ conductor is very close to its standard value, $g=2$ [1], which corresponds to $\beta = 1$ in Eq.(22). Therefore, we consider d-wave-like nodal superconductivity to be much more consistent with the experimental data, where the calculated value $\beta \simeq 0.85$ is close to its expected value, $\beta = 1$. It is not exactly equal to 1, perhaps, due to slightly deviations of superconductivity in $(\text{TMTSF})_2\text{ClO}_4$ from weak coupling scenario. On the other hand, we consider d-wave-like node-less and s-wave parings as very unlikely due to very low calculated value of $\beta \simeq 0.5$.

4. Conclusion

We have suggested explanation of high values of the upper critical magnetic fields, experimentally observed in $(\text{TMTSF})_2\text{ClO}_4$ conductor [13,14,16], using d-wave-like nodal scenario of superconductivity. On the other hand, we anticipate that, for explanation of very high values of the upper critical magnetic fields [33,34] in a sister compound, $(\text{TMTSF})_2\text{PF}_6$, in mixed superconducting-antiferromagnetic phase [34,35], it is necessary to consider either triplet superconducting pairing [10,23,24] or the coexistence of triplet and singlet superconducting order parameters [36].

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